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1. SUMMARY

The experimental progress on contract NAS 7-7 during the past three months may be summarized as follows:

- a. Six cell assemblies and three additional power supplies were constructed.
- b. Seventeen valid charge-retention tests were conducted.
- c. Cell polarization was obtained for several types of oxygen electrodes as a function of pressure and electrode diameter, and for series operation.
- d. Design was completed for a multicell unit, and fabrication was initiated.
- e. Two cycle life tests were performed.
- f. Preliminary evaluations of potential safety hazards and cell capacity were made.

1.1 Charge Retention

The results of the charge-retention tests indicate that the loss of charge via diffusion of the gases through the asbestos bed is very slow. The tests have also revealed that the cell effectively retains its charge, although under some operating conditions it has not always been able to deliver 100 percent of this charge. The cause for the incomplete discharge is attributed to one or more of several controllable factors (cf. Sec. 7.33).

1.2 Polarization

The oxygen electrode studies have revealed that of all the types which have been examined to date, only one exhibits appreciably less polarization than the platinized nickel currently being used. Several have been found, however, which exhibit polarization characteristics of the same order of magnitude as platinized nickel. The studies have revealed that discharge polarization is marked dependent

on pressure in the range between 0 and 200 psig. The results also established that, for electrode diameters of at least 4 in., the discharge current increases linearly with electrode area.

1.3 Cycle Life Testing

One cycle life test was carried out for 29 days (417 "shallow" cycles of 100 min each) of continuous operation, after which the discharge voltage dropped suddenly to zero. The cause of this drop is currently being investigated. Another test was initiated and is currently in its tenth day of semicontinuous operation, consisting of one "deep" 6-hr cycle per day. The voltage and gas pressures have been found to be the same for each cycle.

1.4 Multicell Unit

The final design of the multicell unit was completed, and machine drawings were prepared. The testing of one element was initiated.

1.5 Miscellaneous

Extreme operating conditions were imposed upon the cell in order to evaluate potential safety hazards. The results of several tests showed that no hazard should exist for the condition in which the two gases become mixed, both at room temperature and at 200°F.

Analysis of several "deep" cycles indicates that the cell capacity is approximately 2 amp-hr/in.² of asbestos bed.

2. STATUS AT START OF SECOND QUARTER

All of the investigations for the past quarter were to have been concerned with the particular cell configuration denoted as Cell "B" in the last Interim Engineering Report (1).

At the start of this quarter, three models of Cell "B" had been constructed. One of these was a low-pressure (10 psig) model with glass ehrlenmeyer flasks as the gas containers. The other two were high-pressure stainless steel models.

A method had been devised for preparing both H_2 and O_2 electrodes which yielded reasonable current densities (10 to 15 ma/cm^2) at room temperature and somewhat higher current densities at elevated temperature.

Two charge-retention tests had been conducted with these models: one at 80°F. and one at 200°F. The maximum pressure in these tests was 60 psig, and the maximum stand time was 19-1/2 hr. No cycle life testing had been conducted.

The facilities initially available for the present program included three constant current cycling units, three miniature recorders, three ovens, and a high-temperature furnace with atmosphere control for preparing various types of electrodes.

3. REVIEW OF OBJECTIVES

The objectives for the present quarter consist of the following four items:

- a. To determine the effect of cell configuration and operating parameters on self-discharge.
- b. To investigate the possibility of reducing the cell polarization by preparation and evaluation of oxygen electrodes.
- c. To obtain data on cell lifetime and the effects of continued cycling on cell performance.
- d. To design and fabricate a multicell prototype.

4. SUMMARY OF EXPERIMENTAL PROGRAM

The initial phase of the experimental program consisted of the construction of six cell assemblies and three power supplies. Four of the assemblies were installed within ovens for elevated-temperature testing, and two were installed in small metal cabinets for room-temperature testing.

With these assemblies, a total of 17 successful charge-retention tests were carried out. The tests were conducted within the temperature range of 70 to 290°F and within the pressure range of 0 to 300 psig. Stand times, except for one special 30-day test, varied between 0 and 72 hr.

The same assemblies were used for a number of polarization studies. The first study consisted of the evaluation of several oxygen electrodes including several grades of porous nickel and porous carbon impregnated with various catalysts, lithiated nickel oxide, and a specially prepared electrode from a battery supplier. The second study comprised the measurement of polarization as a function of pressure. The third and fourth studies consisted of the measurement of polarization for series operation and for larger diameter electrodes.

Two cycle life tests were performed during this quarter. The first consisted of a "shallow" 100-min cycle; i.e., 65 min charge at 108 ma and 35 min discharge at 200 ma. This test was conducted on a continuous basis for 29 days. The second test consisted of a "deep" 6-hr cycle; i.e., 1 hr charge at 1.0 amp and 5 hr discharge at 0.2 amp. One such cycle is conducted per day. The test is currently in its tenth day.

Development of a multicell unit was begun in the middle of the past quarter and continued until the present. Progress has proceeded to the point of the preparation of a final machine drawing and the testing of one element.

Additional tests were concerned with the evaluation of the safety hazards involved in the operation of this cell.

5. EXPERIMENTAL TECHNIQUES

5.1 Preparation of Platinized Nickel Electrodes

The platinized nickel electrodes were prepared by immersion plating of the porous nickel electrode in a 3 percent solution of chloroplatinic acid. The porous nickel, 0.022 in. thick, was obtained from Gould National Batteries and was designated as "raw unimpregnated plates." The amount of solution was fixed on the basis of depositing 40 mg of platinum per square inch of electrode.

5.2 Preparation of Carbon Electrodes

Three types of carbon electrodes have been employed in the oxygen electrode tests. The properties of these carbons are listed below:

<u>Type</u> ¹	<u>Porosity, %</u>	<u>Avg. Pore Size, in.</u>
A	40	0.0027
B	60	0.0055
C	36	3×10^{-5} to 4×10^{-4} (range)

The first step in the preparation of these electrodes was the machining of the proper size electrode, 1-5/8-in. dia., from the sample block. The next step consisted of boiling the electrode in distilled water for 1 hr so as to rid the electrode of loose particles of carbon resulting from the machining step. Each of the electrodes was given the special treatments described below, and then each was evaluated as an oxygen electrode.

- a. Platinum black was applied to type A, according to the method described by Hunger (2). Briefly, the method consists of brushing a solution of chloroplatinic acid

¹Type A and B were obtained from the National Carbon Co. and type C from the Speer Carbon Co.

on the electrode and then firing in air to decompose this compound and form platinum black. The amount of solution was based on the deposition of 40 mg of platinum per square inch.

- b. Platinum black was applied to type A by spreading a moistened slurry of the black over one surface. Approximately 100 mg was added per square inch.
- c. Types A and B were impregnated with a mixture of oxides (spinel), according to Kordes (3). The particular spinel contained cobalt, aluminum, and silver oxide.
- d. Type C was activated according to Hunger (2). Briefly, this method consisted of an initial treatment with 50 percent nitric acid, followed by heating for 3 hr at 800°C in a CO₂ atmosphere, as shown in Fig. 1.
- e. Type C was given the same treatment as in the preceding step and in addition, was coated with platinum black, according to Step 1.

5.3 Preparation of Lithiated Nickel Oxide Electrodes

The lithiated nickel oxide electrodes were prepared from the same type of porous nickel as the platinized nickel electrodes described in Sec. 5.1. In this case, however, the thickness was 0.089 in. The lithiating treatment consisted of soaking in a lithium hydroxide solution (11.15 gmc LiOH/100 ml) and then firing in air at 750°C for 5 min.

5.4 Polarization Measurements

The total cell polarization was obtained by use of the constant current power supply. The procedure consisted of setting the current to a given value for either charge or discharge and then measuring the cell voltage on a recorder. After each change in the current setting, a time interval of approximately 3 min was required

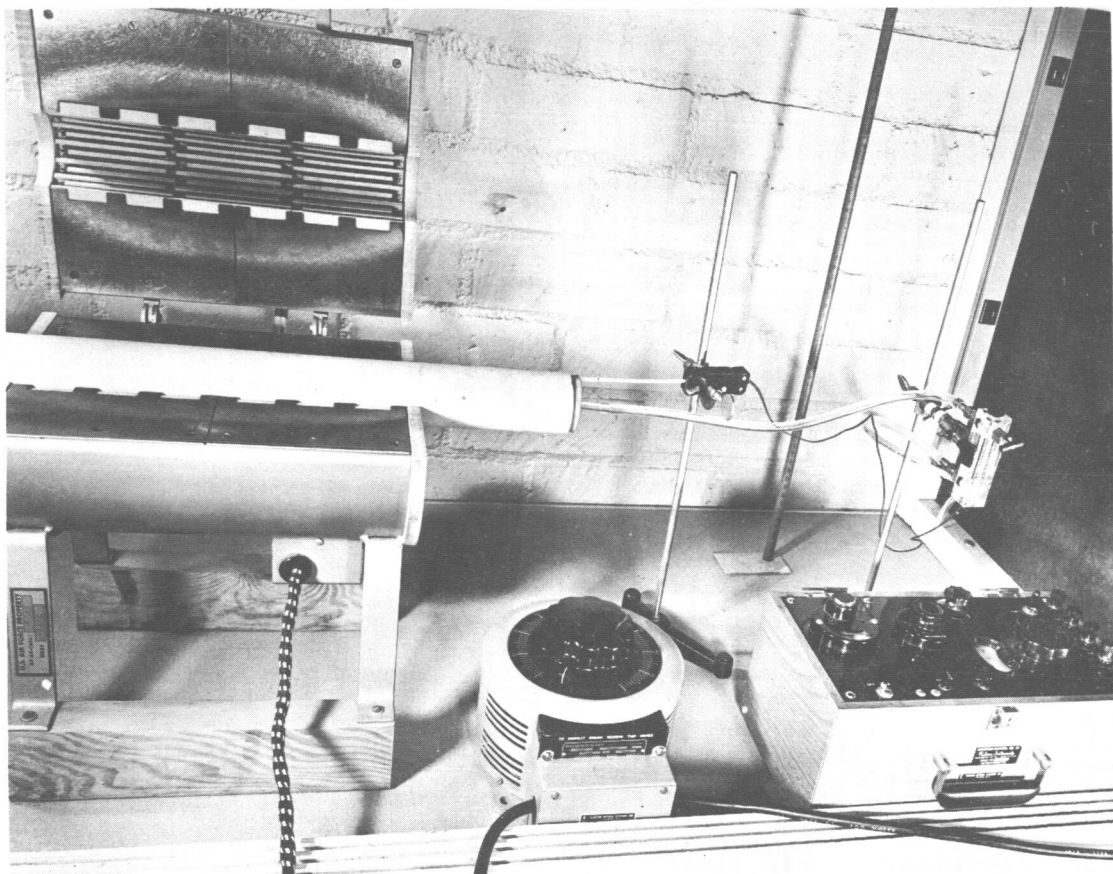


FIG. 1 CONTROLLED ATMOSPHERE FURNACE FOR ELECTRODE PREPARATION

before the cell voltage reached a steady-state value. Several current settings were made to correspond to current densities between 0 and 300 ma/cm².

5.5 Charge Retention

The charge-retention tests consisted of three steps. First, the cell was charged at a constant current of 1.4 to 1.6 amp for a given time interval. Next, the cell was allowed to stand on open circuit for a given time interval (up to 72 hr) at a constant temperature. Finally, the cell was discharged at 0.2 amp until the end of discharge (the time at which the voltage began to fall rapidly toward zero).

5.6 Temperature Measurements

The cell temperatures were measured with an iron-constant thermocouple inserted into a small hole in the cell body. The readings were taken with a Rubicon potentiometer.

5.7 Pressure Measurements

The H₂ and O₂ pressures were taken from precision pressure gauges ($\pm 1/2$ percent of full scale) which were connected to each of the miniature gas cylinders.

6. DESCRIPTION OF EXPERIMENTAL CELLS

The components of one of the cell assemblies are shown in Fig. 2. A brief description of these components is given below:

- a. Hydrogen container: 2-in. dia. stainless steel pipe with welded caps.
- b. Oxygen container: same as hydrogen container, except that its volume is 1/2 that of the hydrogen container.
- c. Cell frames: 4-in. dia. stainless steel, recessed for electrodes and O-ring.
- d. Hydrogen electrode: 1-5/8-in. dia., 0.022-in.-thick disc of wire-reinforced sintered nickel, coated with platinum black at 40 mg/in.²
- e. Oxygen electrode: same as hydrogen electrode except where noted.
- f. Pressure gauges: 0-100 psig, 0-300 psig, and 0-600 psig.
- g. Electrolyte: 35 percent KOH solutions impregnated in a disc of asbestos 2-1/2 in. dia. by 1/16 in. thick.

The six assemblies are shown in Fig. 3. The components of each assembly are identical except for the volumes of the gas cylinders. The volumes are tabulated below, along with the capacity per unit of pressure at room temperature.

<u>Assembly</u>	<u>Hydrogen Volume</u>	<u>Oxygen Volume</u>	<u>Capacity amp min/psi at 70°F.</u>
A	270	135	2.38
B	187	94	1.67
C	150	75	1.32
D	140	70	1.24

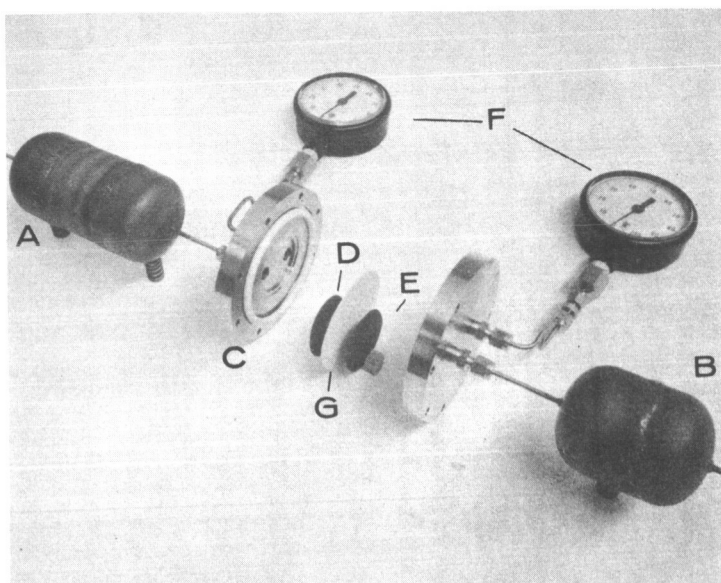


FIG. 2

COMPONENTS OF CELL

- A. Hydrogen container
- B. Oxygen container
- C. Cell frame with "O" ring
- D. Hydrogen electrode
- E. Oxygen electrode
- F. Pressure gages
- G. Asbestos bed

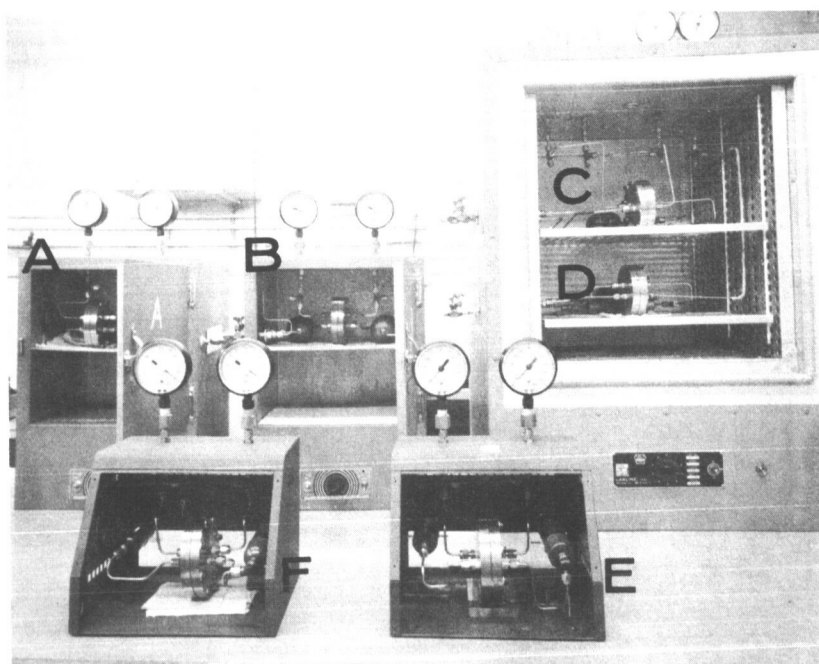


FIG. 3

SIX CELL ASSEMBLIES

<u>Assembly</u>	Hydrogen <u>Volume</u>	Oxygen <u>Volume</u>	Capacity <u>amp min/psi at 70°F.</u>
E	76	38	0.66
F	76	38	0.66

As shown in Fig. 3, four of the assemblies are assembled in ovens for elevated-temperature testing and two in small metal cabinets for room-temperature testing.

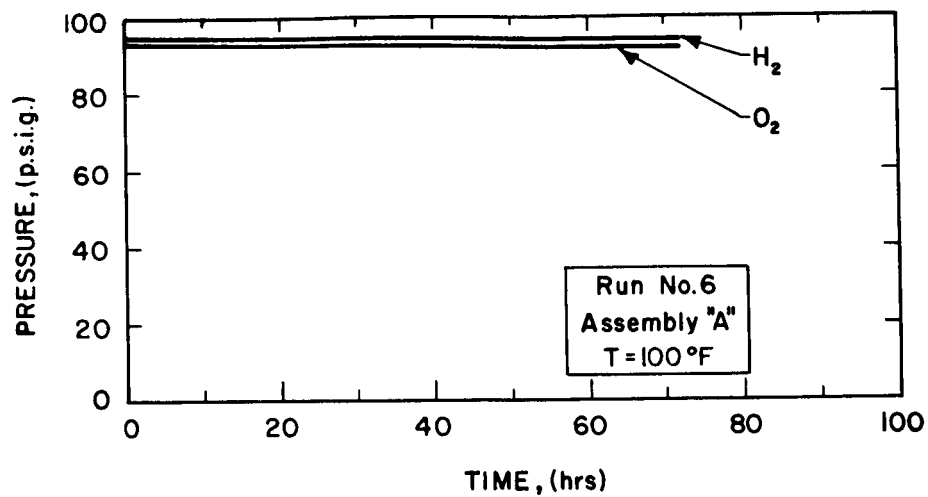


FIG. 4 CHARGE RETENTION AT 100°F

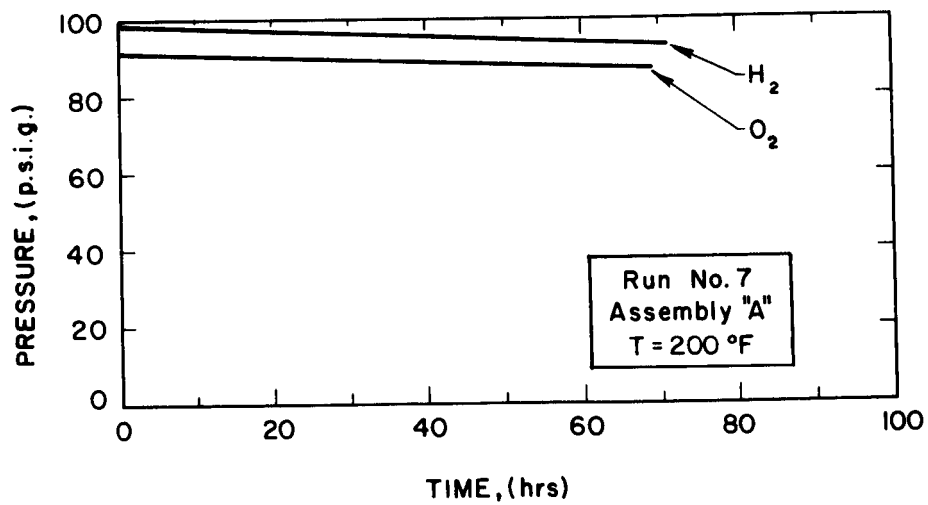


FIG. 5 CHARGE RETENTION AT 200°F

7. RESULTS AND DISCUSSION

7.1 Charge Retention

The state of charge of the regenerative H_2 - O_2 cell can be determined directly at any time by measurement of the two gas pressures.¹ A record of the variation of these pressures with time then defines the charge-retention characteristics of this cell.

If external gas leaks do not occur, a drop in the gas pressures signifies a chemical recombination of the two gases. This recombination occurs via diffusion of the gases through the asbestos bed and subsequent thermochemical reaction to form water.

The time variations of H_2 and O_2 pressures for three sample tests are shown in Figs. 4, 5, and 6. During these particular tests, external gas leaks were either non-existent or so small as to be undetectable by extensive leak testing. (In a number of tests, gas leaks developed in the tubing external to the cells. Data from such tests are considered unreliable.)

Inspection of the charge-retention curves reveals that the drop in gas pressures is very small for a 72-hr period. Figure 4, for example, shows that in Run No. 6, Assembly "A" lost only 1 psi for both H_2 and O_2 after a 72-hr stand at $100^\circ F$. With initial pressures near 95 psig, the percentage loss per unit time becomes 0.015 %/hr. At $200^\circ F$, as shown in Fig. 5, the corresponding loss was found to be 0.075 %/hr.

The results obtained in a still active 30-days charge-retention test at room temperature are shown in Fig. 6. The conditions for this test were specified in order to obtain charge-retention data which may be compared with that of other batteries subjected to the same conditions. Inspection of Fig. 6 reveals that after the first

¹This statement holds, of course, only under the condition that the cell can be discharged completely.

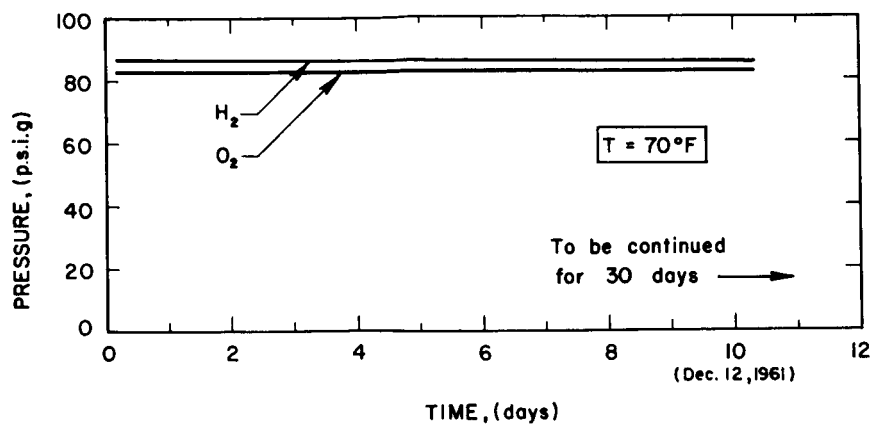
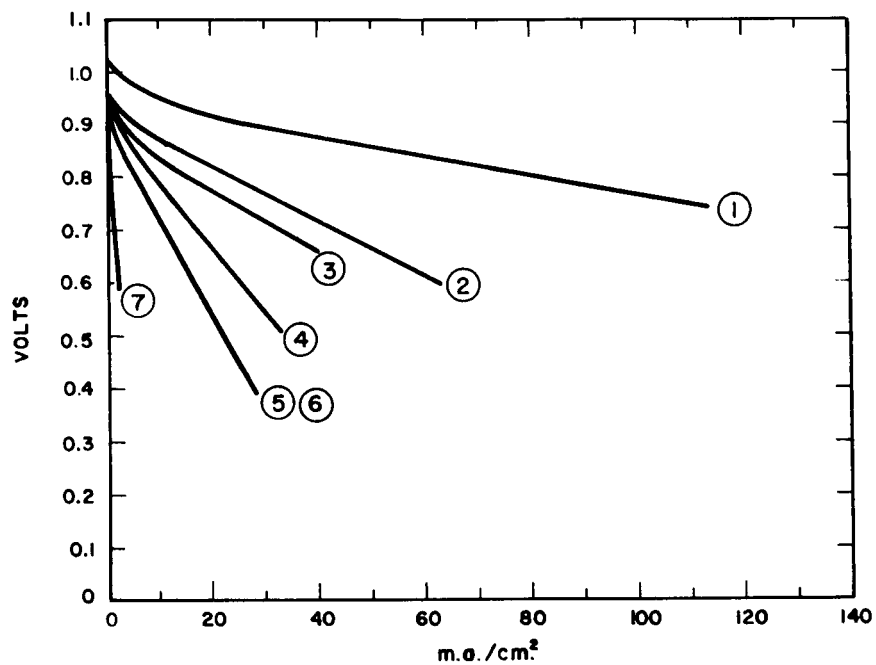


FIG. 6 CHARGE RETENTION AT ROOM TEMPERATURE



- | | |
|---|---|
| ① Special electrode from battery supplier: 30 psig, 150°F | ⑤ Platinized carbon (by decomposition) 30 psig, 200°F |
| ② Platinized Ni: 30 psig, 200°F | ⑥ Platinized Ni: 30 psig, 80°F |
| ③ Lithiated NiO: 30 psig, 200°F | ⑦ Lithiated NiO: 30 psig, 80°F |
| ④ Platinized carbon (by mechanical application) 30 psig, 80°F | |

FIG. 7 OXYGEN ELECTRODE EVALUATION

10 days, the cell has lost approximately 1 psi, or slightly over 1 percent, of the initial pressure. The open circuit voltage has remained constant at 1.02 volts. The cell will be discharged at the end of the thirtieth day.

Although the pressure-time data reveal that the cell can retain its charge effectively, tests show that in some operating conditions, the cell cannot always deliver 100 percent of this charge. An example of this phenomenon is given by Run No. 6 in Table I. As mentioned above, in this particular run, the cell retained approximately 99 percent of the charge after a 72-hr stand. On discharge, however, the cell delivered only 88 percent of the original input. The difference between the retained charge and the output must then correspond to a residual charge. Evidence for the existence of this residual charge is given by the residual gas pressures, as shown in this table. Further inspection reveals the presence of residual charges, varying in magnitude, for all of the runs reported here.

The cause for the incomplete discharge is attributed to one or more of three controllable factors (none of which was completely controlled in these tests). These factors are flooding, pressure differentials, and deterioration of platinized nickel oxygen electrode. A more complete discussion of these factors is presented in Sec. 7.3.3. When complete control over all of these factors is maintained, then the cell should deliver all of the charge which is not lost by thermochemical recombination.

Because of incomplete discharge, the data in Table I cannot be used to correlate current efficiency with temperature, pressure, and stand time. By the end of the next quarter, however, it is anticipated that the conditions necessary for obtaining complete discharge will have been established, and sufficient charge retention data will be obtained to make the above correlations.¹

¹Recent investigations (Dec. 1961) have established that the tolerable pressure differential is approximately 10 psi; i.e., the discharge voltage will not drop to zero until the state of complete discharge if the difference between H_2 and O_2 pressure is not greater than 10 psi. Investigation of the flooding problem is currently in progress. The object of this investigation is to establish the maximum moisture content of the asbestos bed above which the discharge voltage is stable at a reasonable current density of, say, 20 ma/cm².

TABLE I
CHARGE-RETENTION DATA

Run No.	Assembly	Temp. °F	Input amp-hr	Press. Max.* psi	Stand hr	Output amp-hr	ε** I %	Press. After Discharge* psig
1	D	80	2.16	100	14	1.74	81	15
2	C	80	1.16	90	0	1.10	95	5
3	C	80	2.83	150	0	2.69	95	22
4	E	80	2.00	180	0	1.64	83	
5	E	80	2.00	190	0	1.80	90	15
6	A	85	3.20	90	0	3.20	99	1
7	B	88	4.40	175	24	3.33	76	37
8	A	100	3.60	95	72	3.17	88	6
9	A	158	2.80	90	0	2.53	91	11
10	A	159	2.80	100	22	2.60	93	12
11	A	200	2.45	95	72	1.53	63	25
12	C	237	2.00	130	16	1.04	52	80
13	A	250	2.66	95	0	1.32	50	40
14	-	254	1.73	95	0	1.01	59	60
15	C	257	3.00	230	0	1.90	63	12
16	D	257	2.90	210	0	2.25	78	60
17	C	268	1.47	90	0	0.93	63	50

* Since the H₂ and O₂ pressures are nearly equal, only one value is reported here.

** Percent current efficiency.

7.2 Polarization Studies

7.2.1 Oxygen Electrode Evaluation

The object of the oxygen electrode studies was to find an electrode which exhibits less polarization than the currently employed platinized nickel. Another object was to have an alternative oxygen electrode available in case it is found that the platinized nickel electrode deteriorates after prolonged use at elevated temperature.

The evaluation consisted of the measurement of total cell polarization with the various oxygen electrodes. Since the same hydrogen electrode, platinized nickel, was used in all of the tests, the differences in cell polarization can be attributed to differences in the oxygen electrodes.

Figure 7 shows the cell polarization curves as a function of the various types of oxygen electrodes. Inspection of this figure reveals (curve 1) that the polarization for the electrode specially prepared by a battery manufacturer is much less than that of all the others investigated to date. More information on this electrode will be presented at a later date.

The cell polarization at room temperature with the platinized nickel (cf. Sec. 5.1) electrode (curve 6) is by comparison much greater than the polarization obtained with the special electrode and somewhat greater than was obtained with the electrodes described below. Moreover, the polarization of this same electrode at 200°F (curve 2) is markedly less than at room temperature. A complete study of cell polarization versus temperature for this particular electrode will be made in the near future.

The lithiated nickel oxide electrode (cf. Sec. 5.3) was found to exhibit much less polarization at 200°F (curve 3) than at room temperature (curve 7). This phenomenon is undoubtedly due to the semiconducting properties of this material. (This electrode may turn out to be the best as far as stability is concerned (Ref. 4) for continued operation at elevated temperature.)

The polarization of the two best carbon electrodes is shown in curves 5 and 4. The former consisted of a carbon of 60 percent porosity, 0.0055-in.-dia. pore, with a coating of platinum black which was deposited by decomposition of chloroplatinic acid (cf. Sec. 5.2a). The latter was the same type of carbon with a mechanical application of platinum black.(cf. Sec. 5.2b). The polarization curves of all the other carbon electrodes listed below fell within the range of curves 6 and 7.

- a. 60 percent porosity, 0.0055-in.-dia. pore + spinels (Sec. 5.2c)
- b. 40 percent porosity, 0.0027-in.-dia. pore + spinels (Sec. 5.2c)
- c. 36 percent porosity, 0.00003-in.-dia. pore unactivated (Sec. 5.2)
- d. 36 percent porosity, 0.00003-in.-dia. pore activated (Sec. 5.2d)

7.2.2 Effect of Pressure on Polarization

During the course of the charge-retention tests, the observation was made that the discharge voltage appeared to be sensitive to the gas pressure. Consequently, an examination of this relationship was undertaken.

Figure 9 gives the room-temperature polarization for both charge and discharge at three different pressures. Inspection of this figure reveals that the discharge voltage is markedly dependent upon pressure. For example, if the pressure is increased from 50 to 200 psig, the discharge voltage increases by approximately 0.1 volts at 20 ma/cm². The charge voltage is also found to be dependent upon pressure, but in this case, the effect is not as pronounced.

7.2.3 Series Polarization

In preparation for the operation of a multicell unit, two of the assemblies, B and E, were operated in series. The individual as well as the series combined polarization of the units was measured at room temperature. The polarization curves for this mode of operation are shown

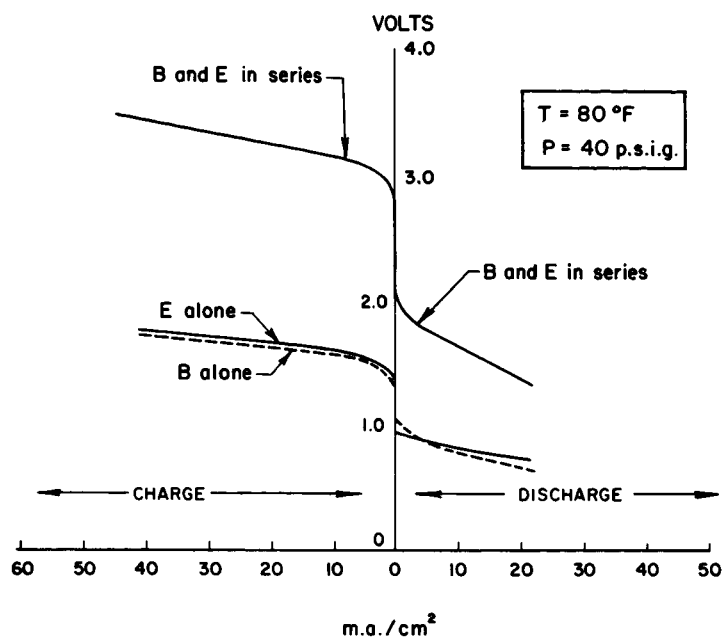


FIG. 8 SERIES POLARIZATION

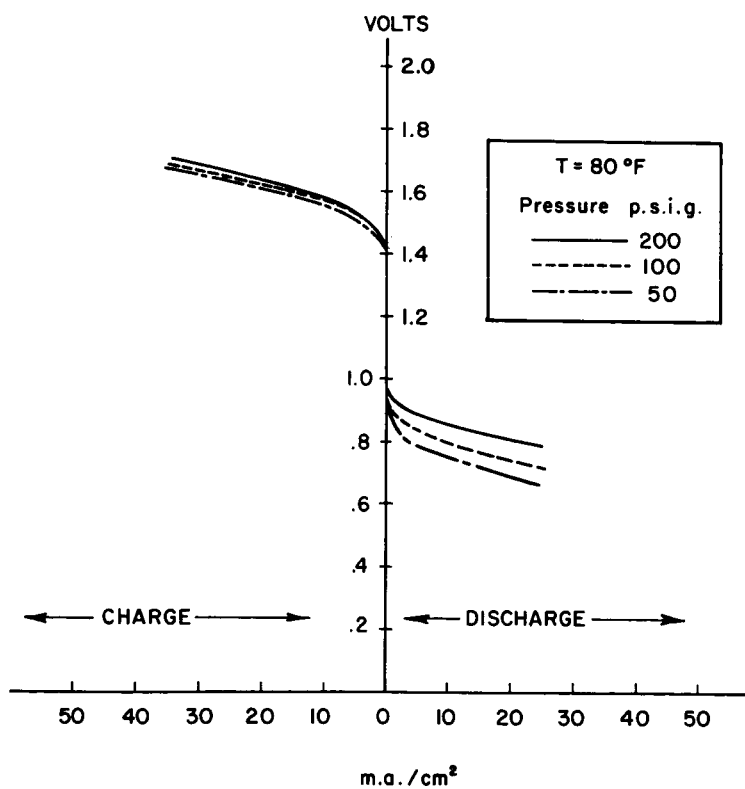


FIG. 9 EFFECT OF PRESSURE ON POLARIZATION

in Fig. 8. Examination of these curves reveals that the series polarization is nearly identical with the sum of the individual cell polarization for the indicated conditions.

7.2.4 Larger Diameter Electrodes

Since the multicell unit is to contain larger diameter electrodes than any of the current laboratory models (4 in. compared to 1-5/8 in.) the polarization of such electrodes was measured in order to ascertain that no adverse effects would be encountered as a result of scaling. Consequently, polarization data were obtained for electrodes of 2-7/16 in. and 4 in. diameter. In each case, the polarization, at a current density of 15 ma/cm^2 , was found to be nearly identical with that of the laboratory model. Hence, the conclusion may be made that the current is proportional to electrode area in the range of electrode diameters employed here.

7.3 Cycle Life Testing

During this reporting period, two cycle life tests were initiated.

7.3.1 65/35 Cycle

Assembly A was placed on automatic cycling at room temperature and initial gas pressures of 50 psig. The cycle consisted of 65 min charge and 35 min discharge (see Fig. 10). The discharge current was held constant at 200 ma, while the charge current was set initially at 108 ma to correspond to a current efficiency of 100 percent. After several days operation, a gradual decline in gas pressures was observed. As a consequence, the charge current was increased to 125 ma and then to 150 ma, at which condition the gas pressures as well as cell voltages remained constant for subsequent cycles.

After 29 days of continuous operation (417 cycles), the voltage dropped suddenly to zero in the middle of a discharge period. Disassembly and examination of the cell revealed no appreciable corrosion of the components. Both platinized nickel electrodes appeared to be the same as when originally assembled, and no contamination of the electrolyte was observed. The weight of the asbestos bed plus electrolyte indicated a slight loss of water.

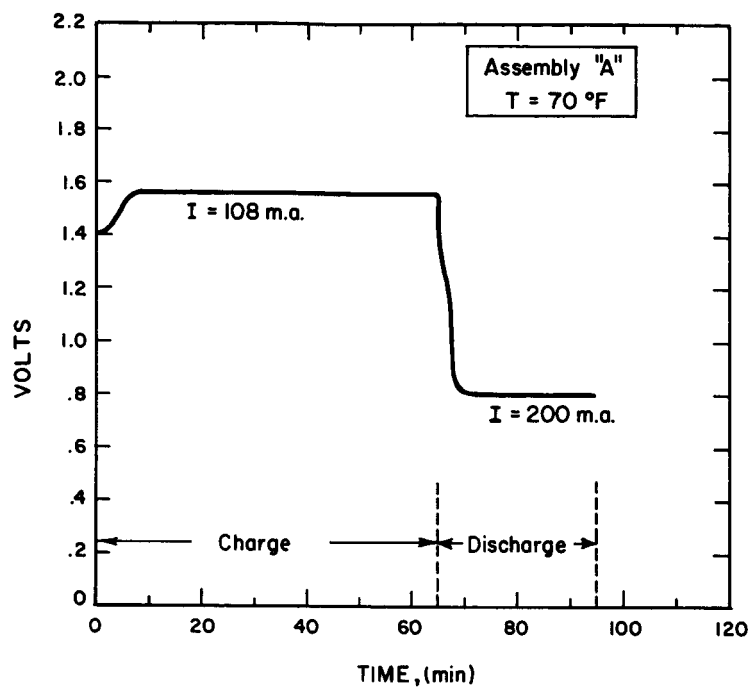


FIG. 10 TYPICAL 65/35 CYCLE

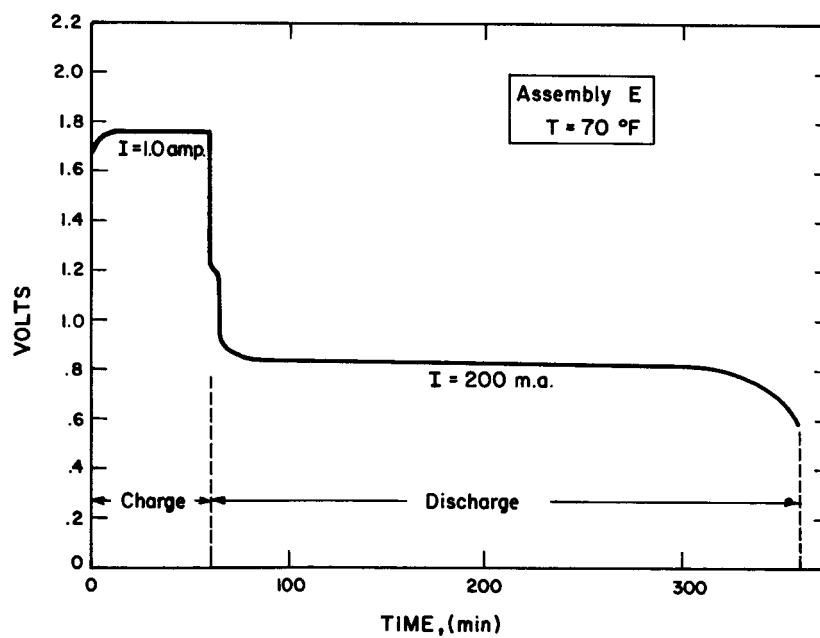


FIG. 11 TYPICAL 60/300 CYCLE

The next step in this test will consist of replacing the old oxygen electrode by a new one and carrying out a few of the above cycles. If the voltage holds for several complete cycles, then the cause for the drop in voltage may most likely be attributed to this electrode.¹

7.3.2 60/300 Cycle

Another room-temperature cycle life test was initiated with assembly "E." The particular cycle (60-min charge at 1.0 amp and 300-min discharge at 0.2 amp) was designed to employ a much greater percentage of the capacity of this assembly than the above test. In this case, the current-time output is 1.0 amp-hr versus 0.12 amp-hr in the above test. Because of this higher output, the maximum pressures, near 160 psig, are much higher. During the course of one complete cycle, the pressures range from 160 psig at the end of the charge to 60 psig at the end of discharge. After 10 complete cycles (see Fig. 11) at 100 percent current efficiency, no drift in either pressure or voltage has been observed.

Since the timing apparatus on the present power supplies has not been set for the 60/300 cycle, the switching has been carried out manually. One cycle, with total cycle time of 6 hr is carried out each day. These cycles will be continued for one month.

7.3.3 Causes of Fall of Voltage on Discharge

Some progress has been made on the determination of the factors that may lead to a fall of cell voltage. However, the problem is not yet completely solved. Apparently, several controllable factors may bring about this phenomenon. With the information obtained to date, the following tentative conclusions are drawn.

¹While this report was in preparation, an inspection of assembly "A," in which this test was conducted, revealed the presence of a short circuit. This short was caused by the metallic contact of the two valves, each of which is connected to one side of the cell and passes through the frame of the oven. The possibility exists then that the fall of voltage was caused by this short.

Flooding

If the electrode becomes flooded with water, then the cell ceases to operate effectively; i.e., the maximum stable discharge current (for several hours) is of the order of only 1 to 2 ma/cm². This fact was demonstrated by intentionally adding an excess of the potassium hydroxide solution to the asbestos bed, charging, and then attempting to discharge at several current settings. At a current setting corresponding to a current density of 30 ma/cm², the voltage fell to zero in less than 1 min. At 20 ma/cm², the voltage fell to zero in approximately 5 min, while at 10 ma/cm², the voltage fell to zero in approximately 15 min. Only at the low current density of 1 to 2 ma/cm² did the discharge voltage remain constant at 0.8 volts. for several hours.

In order to avoid this flooding problem, the amount of electrolyte addition to the bed has been closely controlled in all tests which followed this observation. The amount of electrolyte (35 percent KOH) addition has been established as 0.68 gm solution per 1.0 gm asbestos. With this electrolyte addition, the cell has been shown to operate at the higher current densities for long periods of time (equivalent to 2.0 amp-hr/in.² of bed, as mentioned in Sec. 7.5.2.

High Pressure Differential

If the ratio of the cylinder volumes for H₂ and O₂ is not exactly equal to 2:1, then, when the cell is charged, a pressure differential will develop between the two sides of the cell. This differential will increase as the charging continues until a differential of approximately 15 psig is reached, at which point the gas at the higher pressure is forced through the asbestos bed and into the other gas compartment.

This mixing phenomenon was observed in several of the earlier tests. (The proof of internal mixing was verified by the fact that the pressure of one gas declined while that of the other increased.)

When the conditions were such that a small amount of H_2 was forced into the O_2 compartment, no effect was observed on the discharge characteristics. However, when a small amount of O_2 was forced into the H_2 compartment, then the cell voltage dropped very rapidly on discharge.

The solution to this mixing problem is provided by the use of cylinders with an exact 2:1 ratio for H_2 and O_2 , respectively. Since this may not be realizable in practice, the alternative is to make the ratio as close as possible, with a slight positive tolerance on the 2:1 ratio (i.e., hydrogen volume slightly more than twice oxygen volume). The final adjustment can readily be accomplished in practice by the addition of some inert material to the appropriate cylinder. The method which had been employed at the start of this program was to add water to the cylinder. This procedure was terminated, however, for it was found to cause flooding problems, as indicated above.

The volume of all of the assemblies has now been adjusted so that the pressure differentials are always less than 15 psig (H_2 always greater than O_2) at pressures up to 200 psig.

Oxygen Electrode

During one elevated-temperature test the voltage was found to drop in the middle of the discharge period. Disassembly and examination of the components revealed some corrosion of the O_2 electrode and the stainless steel plate behind it. When this electrode was replaced by a new one, the cell was again found to discharge effectively at constant voltage until complete discharge. The possibility exists that the difficulty was caused by deterioration of the platinized nickel electrode. However, because of the uncertainty of flooding during this test, a proven explanation will require further data.

If the deterioration of this electrode is found to be the cause, then the search for a better oxygen electrode (see Sec. 7.2.1) takes on an additional purpose, i.e., to obtain a more stable electrode in addition to one which exhibits less polarization. More inert materials

may possibly be found in the carbons, lithiated nickel oxide, tungsten, or tantalum.

7.4 Development of Multicell Unit

The development of a multicell unit was initiated during the latter part of October 1961.

7.4.1 Initial Considerations

The basic design of the unit was arrived at by specifying certain performance characteristics and operating conditions, and then calculating the required cell and cylinder sizes. The following design resulted:

- a. Temperature: The operating temperature was selected as 200°F.
- b. Power output: The power output was selected by specifying a discharge current of 3.5 amp at 6.0 volts.
- c. Cell voltage: The discharge voltage was selected as 0.7 volt/cell.
- d. Number of cells: The required number of cells was calculated by dividing the operating voltage, 6 volts, by the unit cell voltage of 0.7 volt per cell. Since the required number was 8.6 cells, 9 cells were chosen.
- e. Electrode area: The electrode area was calculated on the basis of the polarization data at 200°F, i.e., 43 ma/cm² at 0.7 volt. For this current density, it is found that an electrode area of 80 cm² (12.5 in.²) is required to deliver 3.5 amp. The corresponding electrode diameter is found to be 4 in.
- f. Capacity and discharge time: The capacity of this unit is limited by the amount of water which can be electrolyzed from the asbestos bed without affecting

performance. On the basis of the result in Sec. 7.5.3, the maximum capacity is equivalent to 2.0 amp-hr/in.² of asbestos bed. Since the bed area is equal to the electrode area, 12.5 in.², the maximum capacity is found to be 25.0 amp-hr. In order to make a conservative estimate for this first model, the capacity was arbitrarily selected as 1/2 of the above, or 12.5 amp-hr. For a discharge current of 3.5 amp, the corresponding discharge time is found to be 3.6 hr.

- g. Charge time and current: The charge current was arbitrarily selected equal to the discharge current of 3.5 amp. The corresponding charge time is then equal to the ratio of capacity to current, i.e., $12.5/3.5 = 3.6$ hr.
- h. Cylinder sizes: The required sizes of the gas-storage cylinders were determined from the maximum operating pressure which the cell can withstand without performance being affected. Since this pressure has not yet been determined accurately (see Sec. 7.1), the maximum pressure which had been employed successfully in previous tests, i.e., 200 psig, was assumed. The required tank sizes for H₂ and O₂ were then determined by two steps. First, the amount of each gas at 0°C and 14.7 psig was calculated by converting the capacity of 12.5 amp-hr to equivalent amounts of H₂ and O₂. (The resultant gas volumes are multiplied by 9, since there are 9 cells.) The second step consisted in calculating the required gas volumes at 200 psig and 200°F. The resultant values were 4.60 liters for H₂ and 2.30 liters for O₂.

7.4.2 Design of Complete Cell

With the unit cell size, number of cells, and tank volume

specified as above, the next step consisted of the design of the complete cell. The prime emphasis was placed upon compactness and weight in order that the final product might be classified as a flight-weight model.

The design calls for a cylindrical unit with over-all diameter of 6 in. and length of 8 in. The total weight is estimated as 8 lb. The nine cells are to be located in the middle of the cylinder, with the two gas cylinders on either end. The components are held together by six tie bolts equally spaced around the periphery. The cell elements are to be made of nickel-plated lucite (high-temperature grade). Stainless steel weld caps, 4-in. dia., will be used for the gas cylinders. A lighter weight material will be considered for these containers at a later date.

7.4.3 Preliminary Testing

Before proceeding with the construction of the above unit, a considerable amount of preliminary testing has been and will continue to be carried out. As pointed out in Sec. 7.2.3, the first test consisted of the operation of two cells in series. Secondly, polarization data were obtained for larger diameter electrodes as pointed out in Sec. 7.2.4. Thirdly, the construction and testing of one element of the unit shown in Fig. 12 was carried out. The polarization data for this element are shown in Fig. 13, along with the predicted polarization based on the small laboratory units.

This last test constitutes the latest effort in the development of the multicell unit. Subsequent tests will consist of the measurement of polarization data for two elements and cycling of same. Assembly and testing of the nine-cell unit will proceed immediately upon the successful completion of these tests.

7.4.4 Expected Performance

The expected performance characteristics of the nine-cell unit are given in Figs. 14 and 15. These characteristics are all based on the existing performance of the small laboratory assemblies.

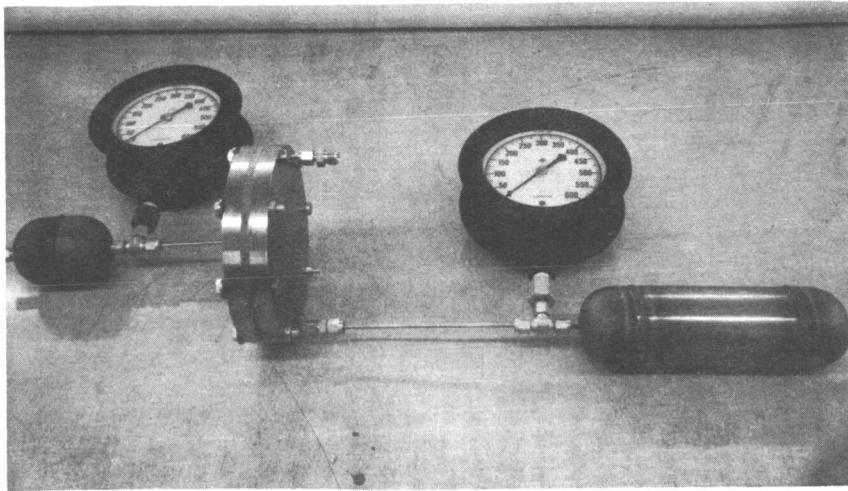


FIG. 12 SINGLE ELEMENT OF 9 CELL UNIT

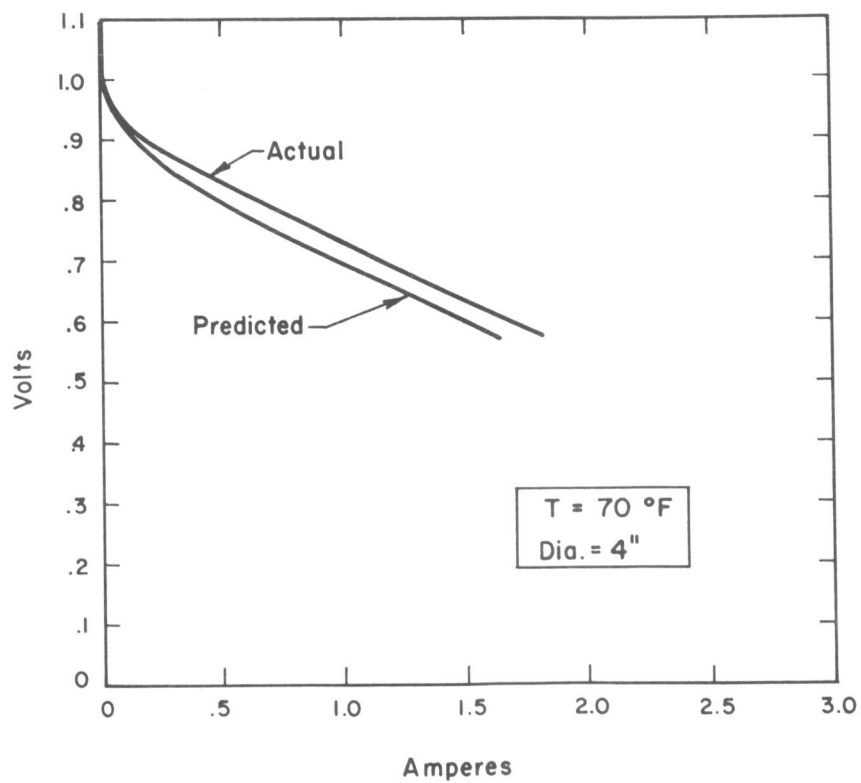


FIG. 13 POLARIZATION OF ONE ELEMENT OF 9 CELL UNIT

Figure 14 gives the expected polarization at two temperatures, and Fig. 15 gives the expected discharge voltage-time curves at the same temperatures.

7.5 Additional Results

7.5.1 Safety Considerations

In order to gain more information concerned with the possible safety hazards involved in the operation of the cells being developed, the following tests were conducted with assembly B containing platinized nickel electrodes.

- a. The cell was charged at room temperature for a period of time such that the H_2 and O_2 pressures were near 100 psig. At this point, the H_2 cylinder was opened to the atmosphere, permitting the H_2 gas to escape and the H_2 pressure to return to 0 psig. Then, the H_2 cylinder was closed. After a few minutes, the O_2 pressure began to decline and the H_2 pressure began to rise, indicating the transfer of O_2 into the H_2 cylinder. After several minutes, the two pressures were nearly equal.

During the whole course of this run no explosion took place.

- b. The same experiment as in "a" was conducted, except that in this case, the O_2 cylinder was opened to the atmosphere. As before, no explosion occurred.
- c. Experiments "a" and "b" above were repeated at 200°F. As before, no explosion was found to occur in either case.

7.5.2 Capacity

Analysis of the results of the charge-retention tests has yielded information on cell capacity. The results show that the cell output is greatly diminished if the cell is charged excessively, i.e., an input greater than approximately 4 amp-hr. For design purposes,

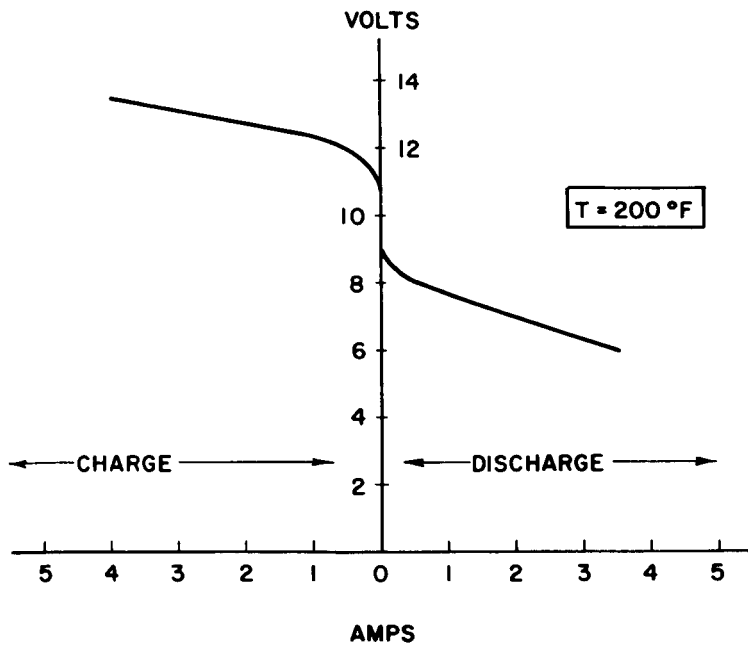


FIG. 14 PREDICTED POLARIZATION OF 9 CELL UNIT

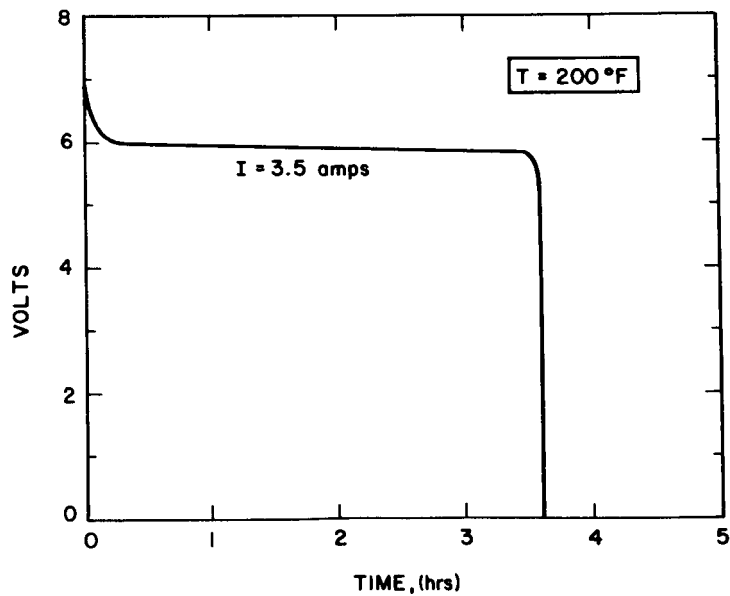


FIG. 15 PREDICTED DISCHARGE VOLTAGE-TIME

this result may be generalized by stating that the capacity is approximately 2 amp-hr per square inch of cell area (electrode area is 2.0 in.²).

The explanation for this "maximum" cell capacity is undoubtedly related to the moisture content of the asbestos bed. Most likely, there exists a minimum moisture content, below which the bed cannot effectively keep the gases separated. This minimum content can be determined quantitatively, for it is known that an electrolysis of 4 amp-hr withdraws 1.3 gm of water from a bed with dry weight of 5 gm which was impregnated with 3.7 gm of 35 percent KOH solution. Based on these values, the minimum content is found to be 0.22 gm H₂O/gm asbestos.

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8. CONCLUSIONS

8.1 Charge Retention

The charge-retention tests have indicated that the loss of charge via mixing of the gases across the asbestos bed is very slow. The tests have also revealed that in spite of the fact that the cell retains its charge effectively, it cannot always deliver 100 percent of the retained charge. The cause of the incomplete discharge is attributed to one or more of several controllable factors (cf. Sec. 7.3.3).

8.2 Polarization Studies

The polarization studies have established that the cell discharge polarization is markedly dependent on pressure between 0 and 200 psig. The oxygen electrode evaluation has revealed that only one of the electrodes examined to date exhibits appreciably less polarization than the currently employed platinized nickel. Several other electrodes have been found, however, which have polarization characteristics of the same order of magnitude. Polarization data on larger diameter electrodes revealed that the discharge current is proportional to electrode area in the diameter range of 1-5/8 to 4 in.

8.3 Cycle Life Tests

During constant current cycling, the cell voltage and gas pressures remain constant from cycle to cycle. The cause for the drop in voltage in one test after over 400 shallow cycles is being investigated.

8.4 Multicell Unit

Based on the results of the testing of one element, the basic design appears feasible.

8.5 Miscellaneous

When a sudden mixing of the gases was brought about by the abrupt release of the pressure in one cylinder, no explosion was found to occur.

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9. FUTURE PROGRAM

9.1 Charge Retention

The charge-retention studies will be continued.

9.2 Polarization

The oxygen electrode studies will be limited to the one or two best electrodes, which will be evaluated extensively. The evaluation will consist of the effect on polarization of the amount of catalyst addition, temperature, pressure, and cycling.

9.3 Cycle Life

The second cycle test now in progress will be continued. Investigations will also be conducted to define more quantitatively the factors which are known to cause a drop in cell voltage, i.e., flooding and gas mixing. Tests will also be carried out to determine whether the deterioration of the platinized nickel oxygen electrode is a cause for the decline of voltage.

9.4 Multicell Unit

The testing of the single element will be completed by carrying out a few "deep" cycles. If the tests are successful, then the construction of the multicell unit will proceed immediately.

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REFERENCES

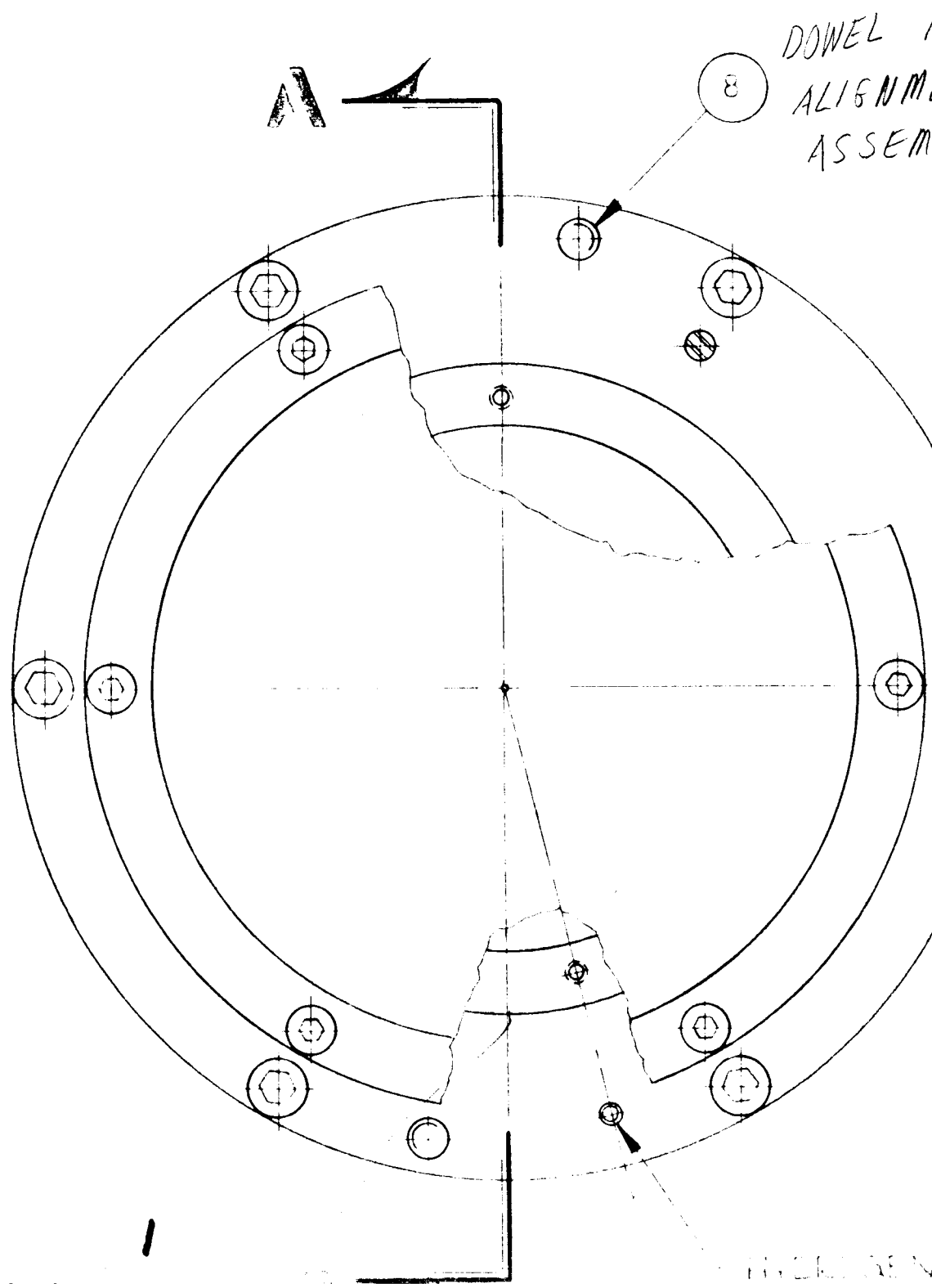
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3. Kordesch, K. "Process for Manufacturing Porous Carbon Electrodes," U.S. Pat. No. 2, 669, 598, 16 February 1954
4. "Proceedings of the Panel on Oxygen Electrodes of the Electro-chemical Working Group," Section 7, published by Power Information Center, University of Pennsylvania, Philadelphia 4, Pa., 28 September 1961

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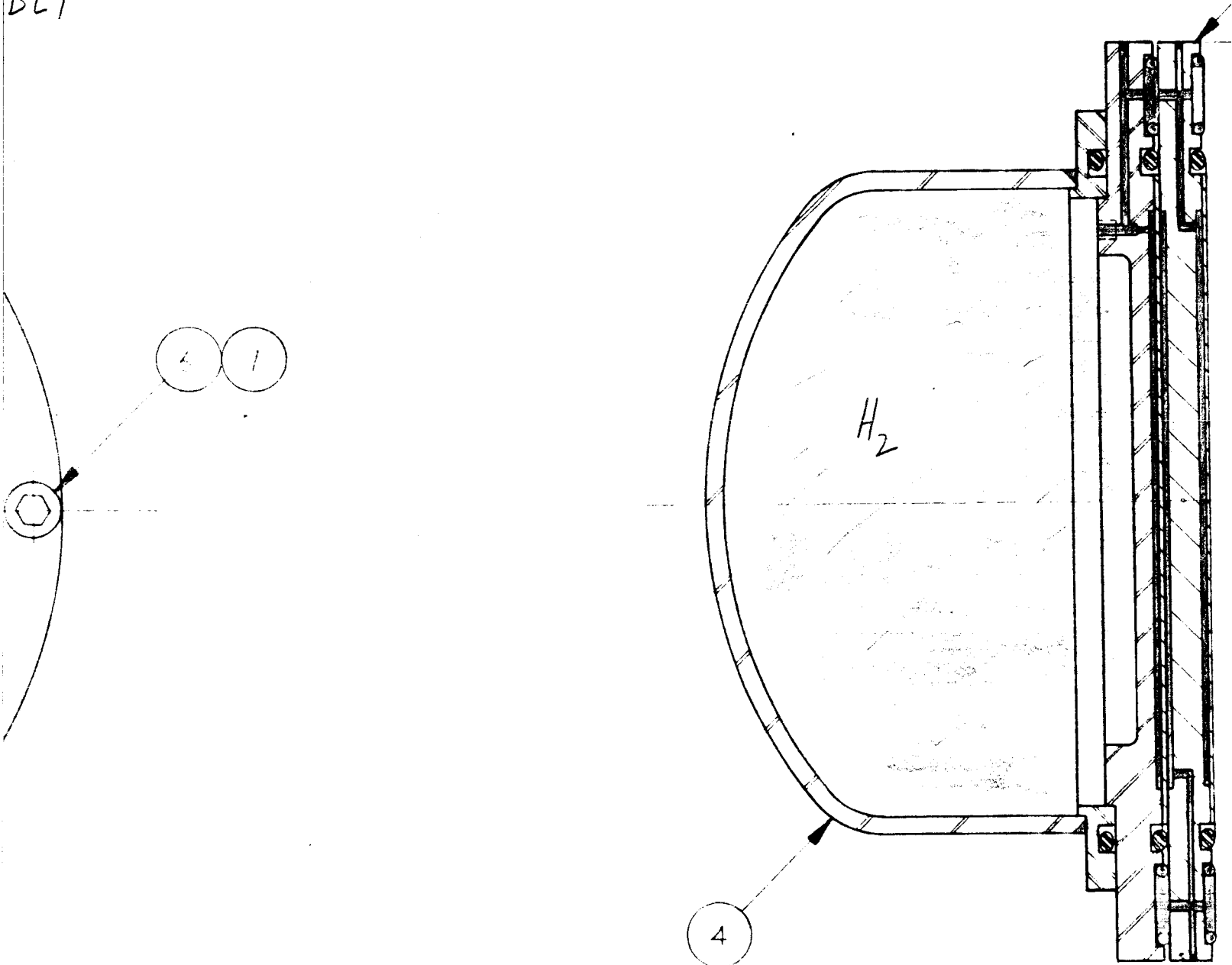
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FIGURE 1

D

Electrodes
H₂: Platinized Nickel
O₂: "
Electrolyte
35% KOH soln in a

PIN FOR
ENT DURING
BLY



C

B

SIZE

DIV.

NUMBER

D

735-

111

022"

Bestor Bed

1 2 3

HYDROGEN PURGE (REF)

Total wt = 9 lbs
No Cells = 9

O₂

5

OXYGEN IN (REF)

2 1/2"

A

REVISIONS

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DESCRIPTION

DATE

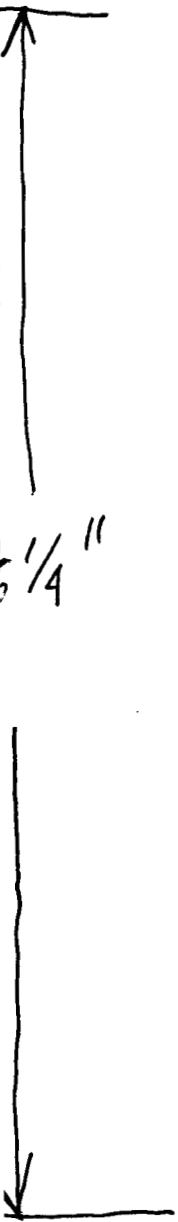
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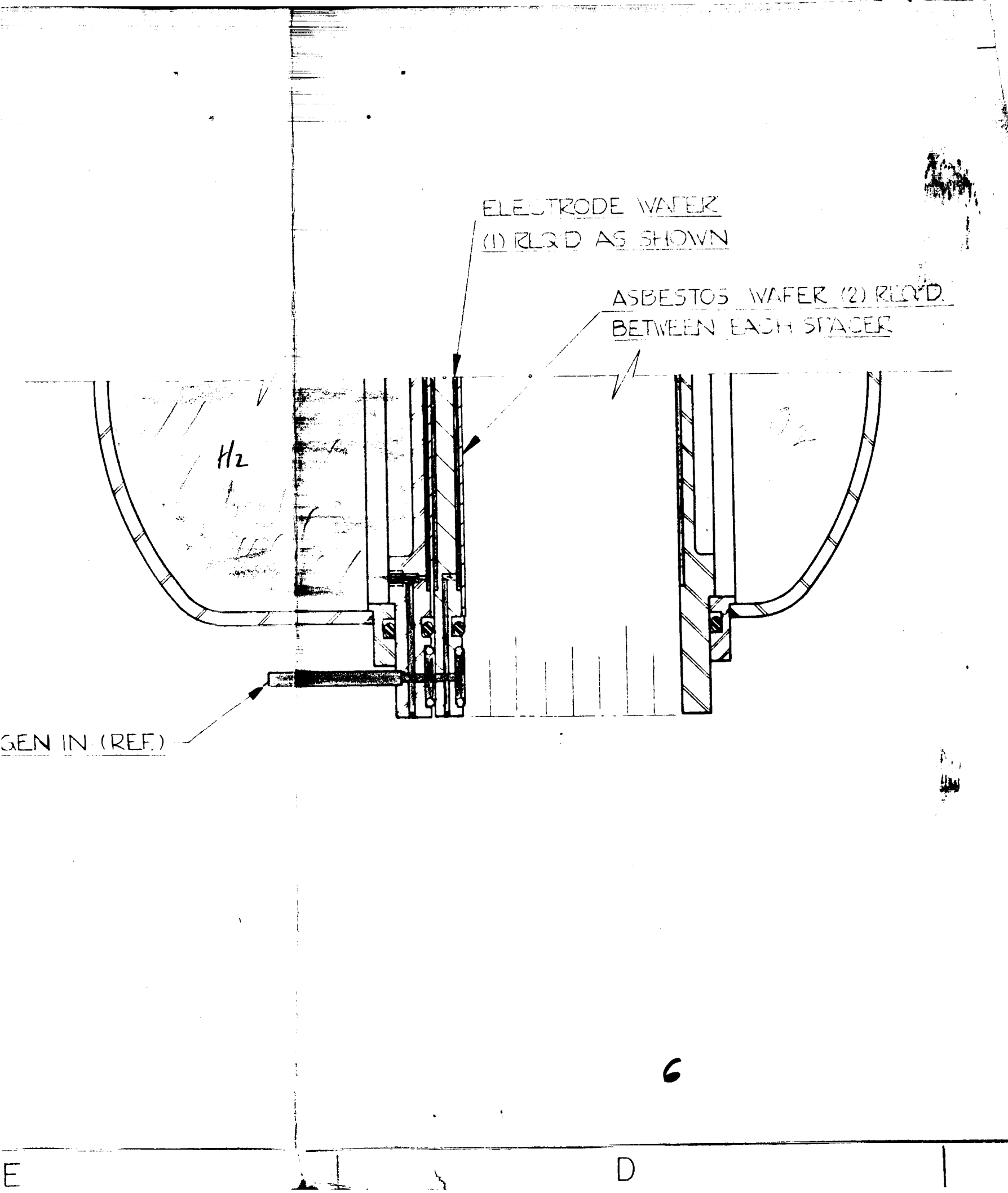
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7

<div>FINAL</div>	<div></div>	SPECIFICATIONS UNLESS OTHERWISE SPECIFIED	
		FRACTIONS	5. FILLET RADIUS 0.10
		DECIMALS	4. SURFACE ROUGHNESS
		ANGULAR	3. THREADS CLASS 2.
		MATERIAL:	2. DIAMETERS TO BE
NEXT ASS'Y.	USED ON	1. REMOVE ALL BURR	
APPLICATION		FINISH	

C

B

8

8		2	1/4 DIA X 3 L5 MICARTA E210
7		6	1/4 -20 HEX NUT
6		6	1/4 -20 X 3 L5, 500, DAP SCR
5	D-11127		TANK C2- END PLATE ATT
4	D-11126	1	TANK P2- END PLATE ATT
3	CAT # 2-110	21	10 PINS 3/16 IN X 3/64 D - HARDEN
2	CAT # 2-116	10	6" L1115 1/8 IN W X 4 4/41 D-ENKEL
1	D-11125	9	SPACER -ELECTRODE
ITEM	PART NO.	REQ'D.	DESCRIPTION

LIST OF MATERIALS

HERWISE NOTED
 MAX
 SS microinch R.M.S. MAX.
 CONCENTRIC WITHIN .005 T.I.R.
 S AND SHARP EDGES.

DRAWN BY

CHECKED BY

APPROVED BY

ARNOLD

HE

TITLE:

FUEL CELL-ASSY.

SCALE FULL

WEIGHT

ELECTRO

OPTICAL

SYSTEMS, INC.

PASADENA - CALIFORNIA

SIZE

DIV

NUMBER

REV

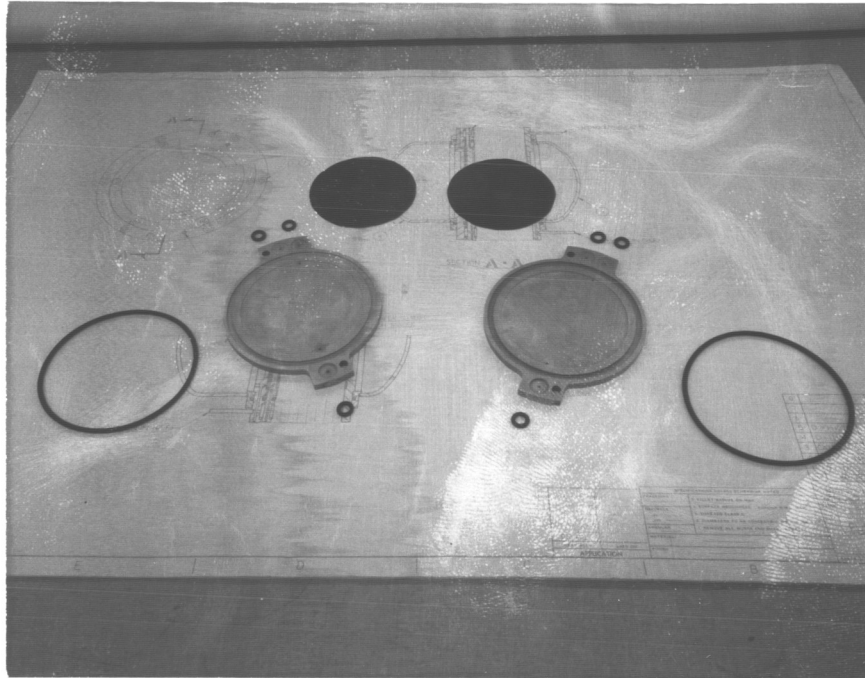
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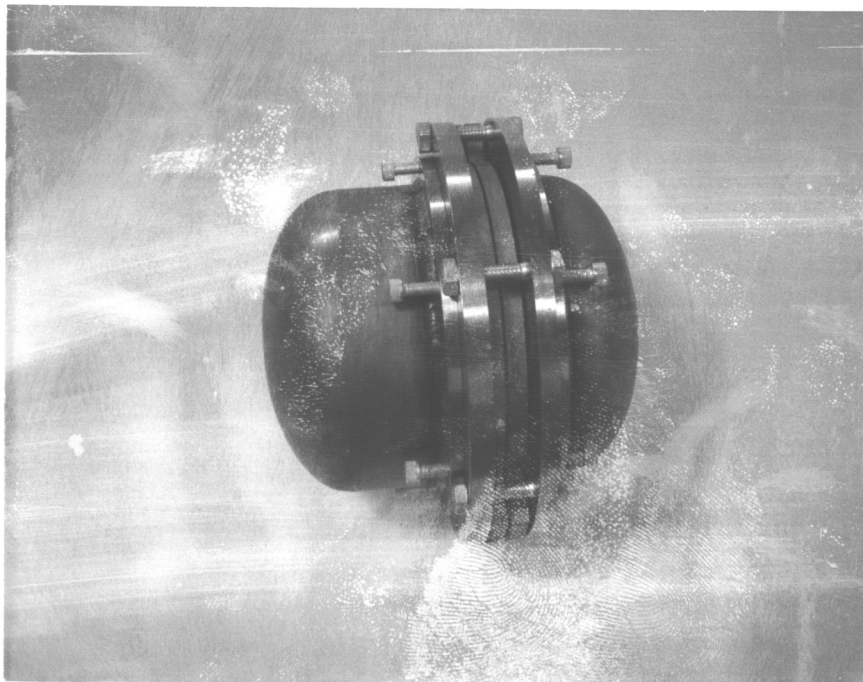
SHEET

OF

COMPONENTS OF MULTI-CELL UNIT



ASSEMBLY OF MULTI-CELL UNIT

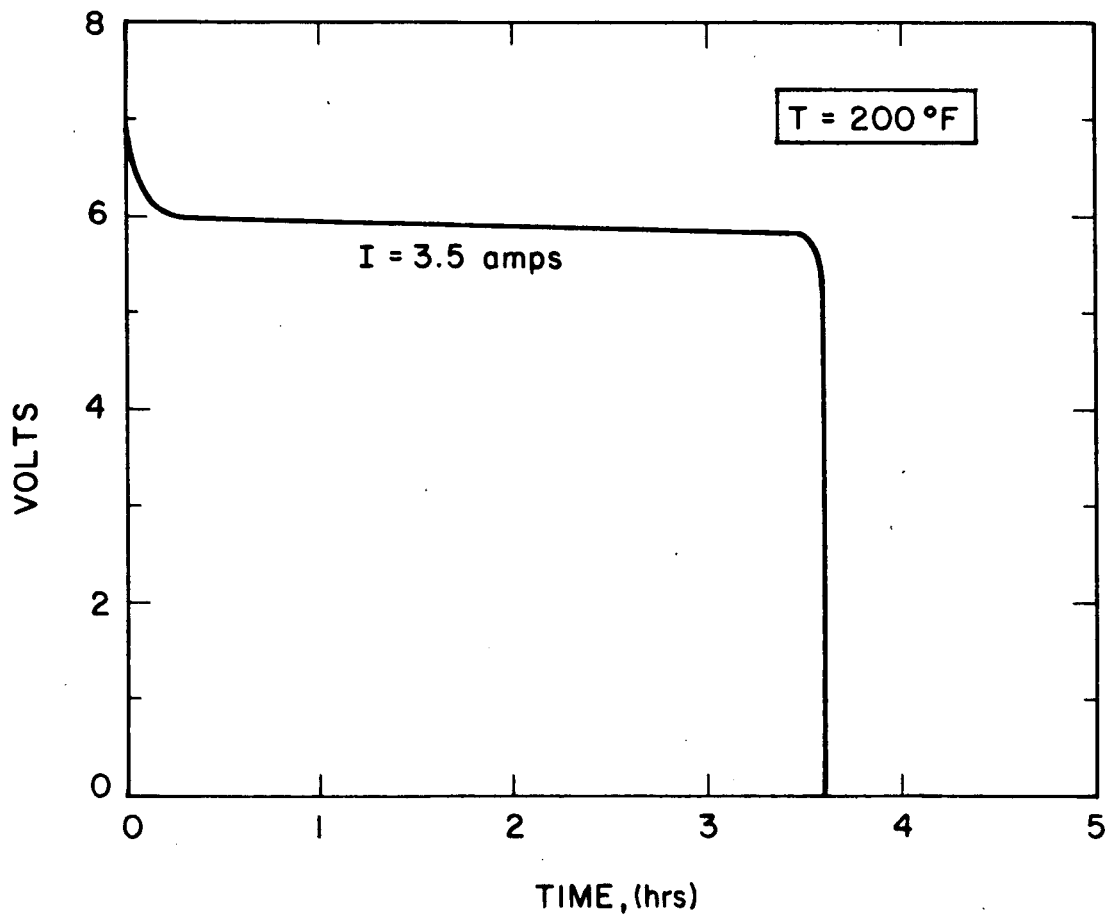


EXPECTED PERFORMANCE CHARACTERISTICS OF
MULTI-CELL UNIT

A.	Operating temperature	70°F to 200°F
B.	Operating pressure	0 to 200 psig
C.	Power output	10 to 20 watts
D.	Charge voltage	11 to 13 volts
E.	Discharge voltage	6 to 8 volts
F.	Charge current	0 to 5 amps
G.	Discharge current	0 to 4 amps
H.	Energy output	70 to 100 watt-hrs
I.	Capacity	12.5 amp-hrs
J.	Weight	9 ^{lbs} hrs
K.	Discharge characteristics	see attached sheet
L.	Polarization characteristics	see attached sheet

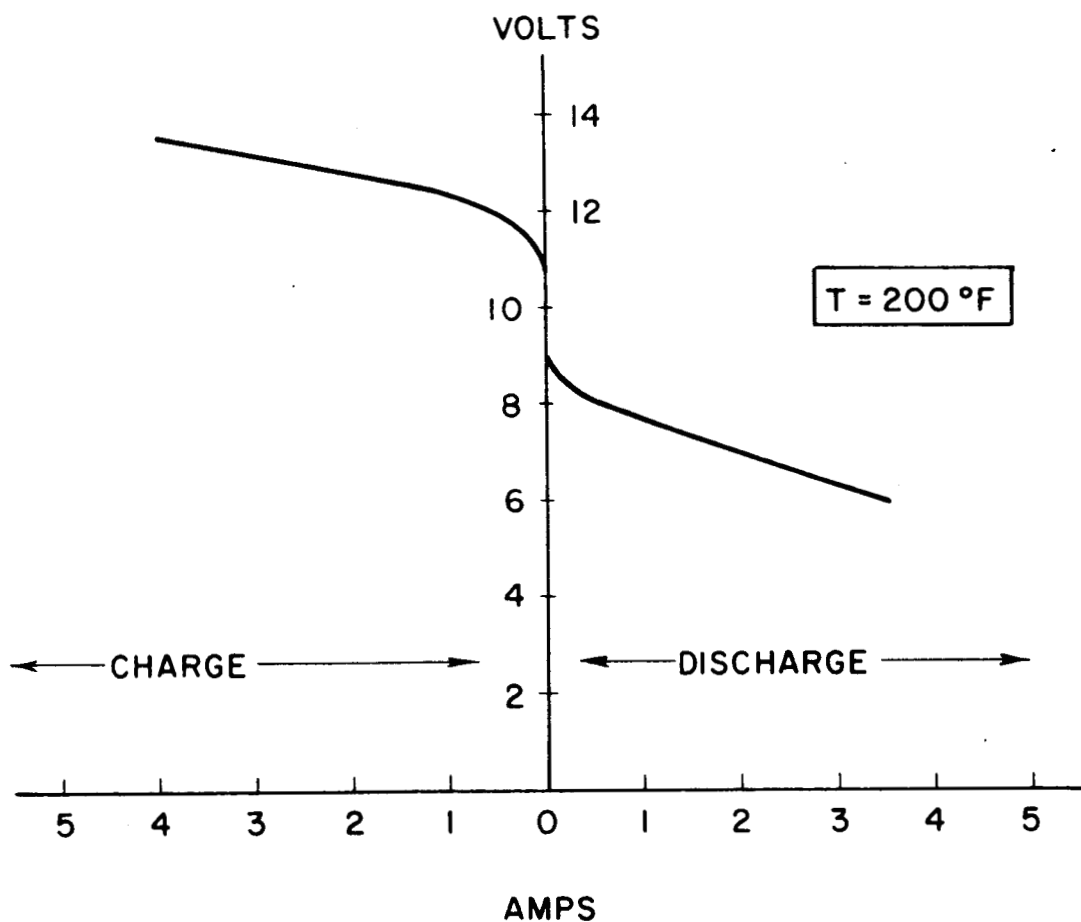
EXPECTED DISCHARGE CHARACTERISTICS
OF 9 CELL UNIT
CHECK PRINT

OK



EXPECTED POLARIZATION
CHARACTERISTICS OF 9
CELL UNIT

OK



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